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February 24, 2016

Analytical Chemistry

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Nuclear Forensics: Scientific analysis supporting law enforcement and nuclear security investigations

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In Nuclear Forensic Science, analytical chemists join forces with nuclear physicists, material scientists, radiochemists, and traditional forensic scientists, as well as experts in nuclear security, nuclear safeguards, law enforcement, and policy development, in an effort to deter nuclear smuggling. Nuclear forensic science, or “nuclear forensics,” aims to answer questions about nuclear material found outside of regulatory control, questions such as ‘where did this material come from?’ and ‘what is the intended use of the material?’ In this Feature, we provide a general overview of nuclear forensics, selecting examples of key “nuclear forensic signatures” which have allowed investigators to determine the identity of unknown nuclear material in real investigations.

On May 29, 1999, a lone male traveler, approximately 35 years old, attempted to leave Bulgaria at the Ruse border crossing, having driven across Bulgaria from Istanbul, Turkey, on his way to his home in Moldova. Bulgarian Customs officers noticed that the traveler fit a “typical smuggler’s profile” – he was alone, he had no baggage other than a small bag that contained his jacket and toiletry articles, he appeared to be overly nervous, and the car he was driving was exceedingly clean. While conducting a secondary inspection, the border guards discovered a document in the traveler’s bag that appeared to be a certificate of analysis with “U235” displayed prominently in the text. A further search of the vehicle uncovered a cylindrical lead container, hidden inside an air compressor. Inside this container was a glass ampoule containing 4 g of a fine black powder (Figure 1). Subsequent analysis of the powder by gamma spectrometry at the Bulgarian Academy of Science determined that the powder contained highly enriched uranium (HEU)¹.

In July 2001, over two years later and almost 1500 miles away, French security services were informed that an individual was trying to sell HEU in Paris, claiming that 30 kg could be made available within a few days. Although the person was a well-known con man, pedestrian teams nevertheless conducted a radiation search of the suspect’s apartment and garage but detected no radioactive material. Information from the security services of a possible meeting involving the suspect at the Place de la Nation in Paris prompted another search. This time a cylindrical lead container was discovered in the suspect’s vehicle, inside of which was a glass ampoule containing 500 mg of a fine black powder (Figure 1)^{2,3}.

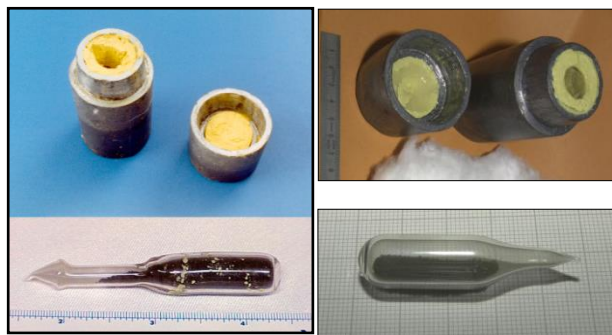


Figure 1. Comparison of the lead containers, glass ampoules, and black powders of the seizures in Ruse, Bulgaria (left) and Paris, France (right) Reprinted with permission from ref 3.

The similarity between the two cylindrical lead containers, including the yellow waxy material apparent in Figure 1, and the two glass ampoules, led to the question as to whether the black powder from these two seizures might be related. Might they have come from the same country, the same facility, the same process, or even the same batch of material? And, if the two materials were related, would that imply that more of the same material might also be outside of regulatory control? These are the types of questions that nuclear forensics seeks to answer^{4,5}.

Extensive nuclear forensic analysis of the two powders was conducted; Lawrence Livermore National Laboratory (LLNL), USA, analyzed the sample seized in Ruse and the Commissariat à l’Energie Atomique (CEA), France, analyzed the sample seized in Paris. The analytical results revealed a remarkable similarity between the two materials. The isotopic composition of the uranium in the two samples was the same (within the expanded uncertainty) and both samples had 2-3 ppb of residual plutonium (see Table 1). This degree of similarity for such an unusual uranium isotopic composition (particularly the high levels of ²³⁶U) suggested a similar origin for the two samples (country, facility and process). However, the ²³⁰Th/²³⁴U model purification dates, as determined by radiochronometry (discussed below), were about one year apart. Such model dates, which correspond to the date that the material was last chemically purified assuming that the

removal of thorium from the uranium was complete at that point, suggested that the two powders were not from the same batch of material.

Table 1. Comparison of analyses of HEU powders seized in Paris and Ruse (U isotopic composition given in atom fraction or atom percent) as reported by CEA and LLNL respectively[#].

	Paris	Ruse
²³² U	9.36E-09%	(1.19±0.72)E-08%
²³³ U	<5.82E-07%	(2.98±0.17)E-07%
²³⁴ U	1.17±0.02%	1.1762±0.0035%
²³⁵ U	72.57±0.86%	72.692±0.027%
²³⁶ U	12.15±0.14%	12.128±0.020%
²³⁸ U	14.11±0.08%	14.004±0.026%
Pu/U	2.20E-09	2.80E-09
²³⁰ Th/ ²³⁴ U	Nov 1994	Nov 1993
Model Date	(±100 days)	(±50 days)

[#]LLNL uncertainties are expressed with a coverage factor of 2 (k=2). The basis of CEA's uncertainties was not described but might reasonably be assumed to be k=2 as well. **Uncertainties for Pu/U are not available.**

Then, in June 2011, almost 10 years after the Paris seizure, authorities in Moldova apprehended a group of middlemen who had attempted to sell 4.4 g of HEU in a cylindrical lead container to an undercover police agent. According to press reports, the group consisted of several citizens of Moldova and the Russian Federation. Moldovan officials believed the material had been smuggled from Russia. The suspects claimed that they could provide between 1 and 9 kg of HEU and that they could also supply plutonium if the buyers demonstrated that they were “serious customers”⁶. Thus far, the analysis of this material has not been reported but the same set of questions raised in the aftermath of the Paris seizure a decade earlier remains relevant.

While nuclear material is strictly safeguarded (highly regulated physical protection requirements and comprehensive accountancy and verification measures), incidents such as those described above have occurred. In the early 1990s, when investigations into incidents of nuclear smuggling were first reported, it was realized that there existed the need to scientifically characterize any nuclear or **other** radioactive materials found outside of regulatory control, determine its processing history and origin and implement measures that would prevent further material being diverted. The International Atomic Energy Agency's Incident and Trafficking Database (ITDB) is a record of incidents in which nuclear and other radioactive material outside of regulatory control is reported by participating states; incidents ranging from illegal possession, attempted sale and smuggling to unauthorized disposal of material and discovery of lost radioactive sources⁷. As of December 2013, the ITDB contained a total of 2477 confirmed incidents, 424 of which involved unauthorized possession and related criminal activities. In the 1993-2013 period, 16 confirmed incidents involved unauthorized possession of highly enriched uranium or plutonium⁷. Clearly this is not a challenge confined to history nor, as the preceding case study may suggest, one limited to European jurisdiction. This feature article provides an overview of nuclear forensics and explores the exploitation of selected nuclear forensic signatures to assist in investigation of such incidents.

What is nuclear forensics?

Nuclear forensics can be defined as “the examination of nuclear or other radioactive materials or of evidence contaminated with radionuclides in the context of international or national law or nuclear security. The analysis of nuclear or other radioactive material seeks to identify what the materials are, how, when, and where the materials were made, and what were their intended uses.”⁸ As such, nuclear forensic practitioners seek to provide scientific results to law enforcement and/or nuclear security personnel and to convey those results, and their technical interpretation, in a way that makes their meaning clear. Further, the scientific findings may be required as evidence in a court of law. For such cases, the measurement results presented in court are likely to be simple properties of the material, properties that potentially prove that the seized material is illegal, such as elemental or chemical composition, sample mass, or uranium enrichment level. Nevertheless, these scientific results, and the laboratory system that produced them, must meet all of the national standards for admissibility of evidence in court (applicable in the country in which the offence is to be prosecuted).

Analytical chemistry underpins many nuclear forensic characterization techniques. But while the analytical methods required to analyze nuclear forensic samples may typically be found in any laboratory that undertakes material characterization, the facilities required to handle and analyze radioactive samples are quite specialized and usually found only in nuclear facilities. Working with nuclear or other radioactive samples requires special precautions; staff that handle and analyze nuclear forensic samples need to be adequately trained in radiation safety and wear appropriate personal protection equipment (PPE).

Material Signatures

In the field of nuclear forensics the term ‘signatures’ describes material characteristics such as isotopic abundances, elemental profile, physical and chemical form **and physical dimensions**, that may be used to link a material, either nuclear or **other** radioactive (non-nuclear, such as those used for medical imaging), to individuals, locations or processes. For nuclear materials (i.e., uranium, thorium or plutonium bearing materials), signatures can be created or destroyed at each step of the nuclear fuel cycle. It is possible for some signatures to carry over through multiple stages of the cycle (for example, signatures present in uranium ore may still be present in a uranium ore concentrate); however, each stage generally produces material with a unique set of sample characteristics (process signatures). Validated signatures measured on known materials of the same type as a sample of interest can assist in the interpretation of analytical data acquired during a nuclear forensic investigation. While measurement of these signatures is an integral part of nuclear forensics, the challenge for nuclear forensics is the far more complex task of identifying the origin of the material by correlating the measured signatures with the sample's history. The correct interpretation of nuclear forensic results is critical for provision of accurate technical conclusions to the investigating authorities. The existence or extent of a database of nuclear or **other** radioactive **material signatures** for comparative purposes can be the limiting factor in a nuclear forensic investigation and the development of such databases or libraries is an ongoing endeavor. Some of the nuclear forensic signatures that have been the most useful for answering the questions of law enforcement to date are discussed below.

Analytical Techniques

Nuclear forensic signature measurement relies on analytical techniques from a wide range of disciplines, particularly analytical chemistry, radiochemistry (i.e., chemistry of radioactive elements and compounds) and material science. Techniques can be broadly

classified into three classes, summarized below in Table 2, which have varied, though complementary and equally important, roles. In nuclear forensics there is no universal ‘process flow’ or analytical plan for each unknown sample that is received by the laboratory. The choice of technique applied to each sample is driven by the investigative questions for which answers are sought and the time that is available to answer those questions. The planned sequence of analytical measurements for any particular sample needs to be flexible; the results of one set of analyses may require that the plan be modified. The IAEA has provided guidance on laboratory methods and techniques to be applied during a nuclear forensic examination with typical timescales for completion of analyses (24 hours, 1 week and 2 months)⁸. All of the many analytical techniques that contribute valuable information during a nuclear forensic investigation cannot be covered in this paper. More detailed technical reviews are available for the interested reader^{9,10}.

TABLE 2. Classes of techniques applied to nuclear forensics

Class of Techniques	Examples	Applications
Bulk analysis	X-ray fluorescence (XRF) and X-ray diffraction (XRD)	Characterize the elemental and isotopic composition of the bulk material
	Inductively coupled plasma mass spectrometry (ICP-MS)	Detect and quantify trace constituents
	Gamma spectrometry	
Imaging	Optical microscopy	Determine sample homogeneity or heterogeneity
	Scanning electron microscopy	Assess material morphology and micro-structure
Microanalysis	Secondary ion mass spectrometry (SIMS)	Quantitatively or semi-quantitatively characterize the individual constituents of the bulk material
	X-ray microanalysis	Particle analysis
		Analyze thin layers or coatings

Physical Dimensions

Measurement of a sample’s physical dimensions is one of the first analyses conducted during a nuclear forensic investigation and though it is simple to perform the results can serve as a powerful means of characterizing an unknown sample; size and shape alone may provide sufficient information for identification.

Physical dimensions are particularly useful in the case of nuclear fuel pellets. Nuclear reactor fuel is usually manufactured by compacting uranium dioxide (UO_2) powder, which may or may not be enriched uranium, to cylindrical pellets, followed by sintering at high temperatures to produce high density ceramic fuel pellets. Each reactor type (e.g. heavy water reactor or pressurized water reactor) uses fuel pellets of specific dimensions. Pellets may have one ‘dished’ end face and may have a central hole, both of which provide a space for gaseous fission products produced during irradiation in the reactor. Such characteristics were observed in a nuclear forensic investigation of uranium fuel pellets by Wallenius et al.¹¹. In June 2003, four uranium pellets from Lithuania were received by the Institute for Transuranium Elements (ITU), Karlsruhe, Germany. The dimensions of the four pellets were similar

and they all had one concave end face and a central hole (Figure 2). The pellet dimensions, as well as the uranium enrichment levels, were sufficient to identify the reactor type; an RBMK-1500, a Russian type water-cooled, graphite-moderated reactor. At the time, there was only one operating RBMK-1500 unit in the world, Ignalina Unit 2 in Lithuania.

Physical dimensions of fuel pellets (i.e. diameter, height and weight) were used to identify the origin and intended use in the nuclear fuel cycle in a case described by Mayer et al.¹² In March 1992, a shipment consisting of 72 uranium pellets was intercepted in Augsburg, Germany. The physical dimensions (together with the isotopic composition of the uranium) revealed the intended use of the material – fuel pellets for a Russian type graphite moderated reactor (RBMK).



Figure 2. Fuel pellet described by Wallenius et al.¹¹ where physical dimensions were a key nuclear forensic signature. Reprinted from ref 11. Copyright 2006 Elsevier.

One branch of nuclear forensics is often called ‘nuclear archaeology’ because it applies the methodologies of nuclear forensics to problems of historical interest. In one such case, the physical dimensions were a clear nuclear forensic signature; a uranium metal cube, referred to as the ‘Heisenberg Cube’, investigated by Mayer et al.¹³ The cube was believed to have been part of the German nuclear development program in the 1940’s. Early nuclear reactor design by German researchers configured the uranium in the form of 664 cubes. At the end of World War II, 659 cubes were recovered but 5 were missing. In 2002 a metallic cube was presented to ITU for nuclear forensic examination. Such a cube shape for 2.4 kg of uranium metal is very distinctive (Figure 3). Age determination (see discussion below) of the uranium composing the cube gave September 1943 (± 0.5 years) supporting the physical measurement data.



Figure 3. Uranium metal cube ('Heisenberg Cube') investigated by Mayer et al.¹³ Reprinted with permission from ref 13. Original published under a CC BY-NC-SA license by IOP Publishing.

Isotopic composition of uranium, plutonium and stable elements

Uranium isotopic composition is an essential signature of uranium material. Uranium ore and uranium ore concentrates (UOC or 'yellow cake') have natural isotopic abundance, in which the ^{235}U abundance is 0.72% and the ^{238}U abundance is 99.27%. Natural uranium also has trace amounts of ^{234}U , typically 53-55 ppm. Following conversion and enrichment, uranium may be depleted (^{235}U abundance less than 0.72%), low enriched (LEU, ^{235}U abundance greater than 0.72% and lower than 20%), or highly enriched (HEU, ^{235}U abundance equal or greater than 20%). Irradiation of the uranium fuel or targets by neutrons in a nuclear reactor produces the isotopes ^{236}U and ^{232}U so the presence of these isotopes in 'unirradiated' uranium materials indicates the material is recycled or mixed with recycled uranium.

Uranium isotopes can be measured using radiation counting techniques (such as gamma spectrometry or alpha spectrometry) or with a higher precision using mass spectrometry (e.g. thermal ionization mass spectrometry, TIMS; inductively coupled plasma mass spectrometry, ICP-MS; accelerator mass spectrometry, AMS; or secondary ion mass spectrometry, SIMS).

Since pre-irradiated nuclear fuel may contain a range of uranium isotopic compositions a close match in the uranium isotopic abundances may serve as a useful nuclear forensic signature for possible production facility and intended reactor type. For example, uranium isotopic abundances were measured on the fuel pellets examined by Wallenius et al.¹¹ and Mayer et al.¹², discussed above, and, along with the physical dimensions, were sufficient to identify the reactor type they were manufactured for.

A nuclear forensic investigation into uranium oxide powders seized in Victoria, Australia, revealed both samples were depleted (Figure 4)¹⁴. These two samples (42.9 g of a bright yellow powder and 48.6 g of a dark green powder) were discovered by police during a raid on an illegal amphetamine laboratory. During the ensuing nuclear forensic investigation, scientists measured the uranium isotopic abundances and found that the samples contained 0.44 and 0.41 at-% ^{235}U respectively and also contained ^{232}U and ^{236}U . Hence, they were able to inform law enforcement authorities that Australia was not the source of the samples, since that country does not have enrichment facilities.



Figure 4. Two samples seized in police raids in Victoria, Australia. The uranium isotopic abundance of the samples revealed that the samples did not originate from Australia¹⁴.

Meyers et al.¹⁵ demonstrated how uranium isotopic analysis on environmental samples can be used for nuclear forensic purposes. Soil samples were collected at two former uranium facilities; at one site uranium was refined and fabricated into metal ingots for fuel or targets, and at the other site uranium was rolled into rods. The authors found that the $^{235}\text{U}/^{238}\text{U}$ signatures from each site were distinct and consistent with the activities and processes conducted at each facility.

Plutonium isotopic composition, which can be measured using either radiation counting or mass spectrometry, may also serve as a useful nuclear forensic signature. Plutonium is produced by neutron reactions with uranium, either naturally at extremely low levels¹⁶, in nuclear weapons (found as fallout from post detonation debris in environmental samples) or, more commonly, in nuclear reactors. An area of current scientific interest for nuclear forensic scientists is the potential for plutonium isotopic ratios measured in spent fuel (especially the ratio of ^{239}Pu to the higher mass isotopes formed from higher fuel burn-up) to correlate with the particular reactor in which the fuel was irradiated. These correlations are complicated by such factors as the neutron energy spectrum and neutron flux in the reactor and the starting uranium enrichment of the fuel⁹.

A 'nuclear archaeology' case which illustrates the forensic use of plutonium isotopic ratios is that of a glass bottle containing plutonium (several hundred milligrams in the form of a white precipitate mixed in ~400 mL of clear liquid) which was discovered in a safe recovered from a waste trench at the Hanford site in Washington State in western United States (see Figure 5)¹⁷. Hanford was the location for U.S. plutonium production during World War II. Plutonium isotopic composition measured on the sample, in combination with reactor burn-up modeling, revealed that the sample originated from the X-10 reactor in Oak Ridge, Tennessee, and was transported to Hanford as part of the first batch of plutonium separated at Hanford's 'T-Plant', the world's first industrial scale reprocessing facility.



Figure 5. Excavated safe (left) and glass bottle containing plutonium (right) recovered from the Hanford site in Washington¹⁷; plutonium isotope ratios were a key signature in the determination of the sample's origin. Reprinted from ref 17. Copyright 2009 American Chemical Society.

The isotopic compositions of stable elements such as strontium, neodymium or oxygen have also been used as nuclear forensic signatures. These elements may be present in uranium samples either from endogenous sources (i.e., from the uranium ore) or introduced during processing throughout the fuel cycle (e.g., trace elements introduced as contaminants in chemical reagents used during processing or metal casting). A UOC sample seized during a police raid of a clandestine drug laboratory in Australia in early 2009 (see Figure 6) provides a good example of where the strontium isotope ratio of the sample helped identify the source of the material¹⁸. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for the seized sample was quite

unique and matched that of a known sample from the former Australian operating mine, Mary Kathleen. Similarly, the lead ratios ($^{208}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$) on the unknown UOC also matched Mary Kathleen and were quite different to a range of other UOC samples sourced from other Australian and international uranium mines.

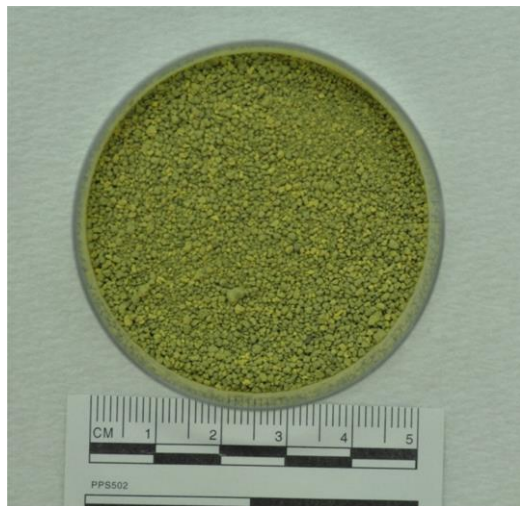


Figure 6. Uranium ore concentrate sample seized in Australia. The Sr, Pb isotope ratios and rare earth element pattern were pivotal for matching the sample to material produced at a former Australian uranium mine¹⁸. Reprinted from ref 18. Copyright 2014 Elsevier.

Trace elemental impurities, anions and REE pattern

Analytical chemists frequently measure trace elemental impurities in various products and this expertise can be directly applied to nuclear forensics where the ‘product’ is nuclear material. As noted above, elemental impurities in uranium materials can be source inherited (i.e., present in the original ore material) or added during processing, either intentionally (for example, gadolinium addition to nuclear fuel to achieve higher burn-up) or unintentionally (such as reaction vessel corrosion products). As uranium is progressively processed to produce nuclear fuel, it is purified, thereby removing most endogenous elemental impurities (the extent of removal depends on the particular impurity element and the particular process). For UOCs though, elemental impurities have proved useful as a nuclear forensic signature for several reported cases¹⁹.

In one such case, nearly 3 kg of radioactive ‘wet brownish yellow powder’ was discovered in a scrap metal shipment in Rotterdam harbor, The Netherlands, in 2003^{12,20}. Scientists from ITU conducted the nuclear forensic examination and found the powder to be an impure uranium compound. While the $^{235}\text{U}/^{238}\text{U}$ and $^{234}\text{U}/^{238}\text{U}$ isotopic ratios were those of natural uranium, the material had clearly been subjected to processing as uranium progeny (e.g. radium and polonium), which are present in the unprocessed uranium ore, were not detected by gamma spectrometry. Elemental impurities were present at relatively high concentrations (>1000 µg/g of material of Al, Ca, Cr, Fe, Mg, Mo, Na, Ni and P). Using these elemental impurity and rare earth element (see below) results, in conjunction with uranium, strontium and lead isotope ratios and other techniques, they were able to identify the material as likely originating from the Middle East.

Anions such as fluoride, chloride or sulphate have been investigated as potential nuclear forensic signatures for UOC samples and are found to be either source related (e.g. high F/Cl ratios were observed in the aqueous leachate of UOCs that had been produced as a by-product from phosphorite ores which typically

contain apatite minerals such as $\text{Ca}(\text{PO}_4)_3\text{F}$ or, more commonly, process related signatures^{21,22}. The use of the $\text{SO}_4^{2-}/\text{Cl}^-$ ratio proved to be useful in the attribution of the UOC seized in Australia, strongly suggesting that the UOC sample had experienced sulphate leaching as part of its processing history (Figure 7)

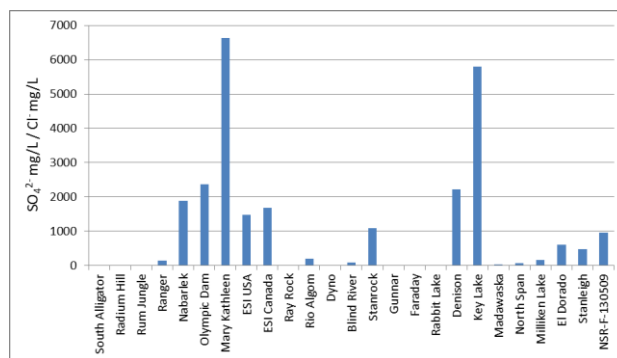


Figure 7 Sulphate/chloride anion ratios of a seized Australian UOC sample (NSR-1-130509) and a range of other UOC samples¹⁸. Reprinted from ref 18. Copyright 2014 Elsevier.

Rare earth elements (REE, i.e. the lanthanide elements) are particularly valuable as nuclear forensic signatures. The pattern of the REEs reflects the geological ore type from which the sample originated i.e., the pattern of concentrations is not altered significantly during processing of the uranium ore due to the similar chemical properties of the REE^{23,24}. For the UOC sample seized in Australia, discussed above, the REE pattern provided compelling evidence that the sample was sourced from Mary Kathleen uranium mine as it was very similar to both a sample of UOC and uranium ore from the suspected mine site (Figures 6 and 8).

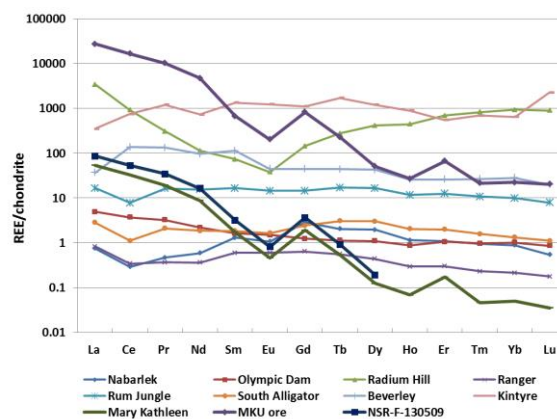


Figure 8. Chondrite normalized rare earth element profile of a sample seized in Australia (NSR-F-130509) exhibited a similar pattern to the uranium ore and UOC sourced from a former uranium mine in Australia¹⁸. Reprinted from ref 18. Copyright 2014 Elsevier.

Age determination

Skills in analytical chemistry and, more particularly radioanalytical chemistry, are essential for the measurement of perhaps the most useful nuclear forensic signature, the material’s age. The age of a sample in the nuclear forensics context refers to the time since the last separation of the progeny isotopes from the radioac-

tive parent (usually uranium or plutonium), referred to as the production (or sometimes separation) date. Separation processes set the clock to 'zero'; the subsequent ingrowth of the progeny isotope allows the age of the sample to be measured (via measurement of progeny to parent ratio) and calculated based on radioactive decay equations²⁵⁻²⁷.

Age determination is an extremely useful nuclear forensic signature as it is non-comparative or 'predictive' i.e., it doesn't require comparison with other material in a database. However, it must be applied carefully. The measured age is referred to as the 'model' age as it relies on a number of assumptions; complete separation of the progeny isotope from the parent at time zero (if the separation is incomplete, the calculated age or 'model' age will be older than the true age) and also that the system is 'closed' i.e., no loss or gain of the parent or progeny other than through radioactive decay of the parent.

The most commonly applied radiochronometer in nuclear forensics is the decay of ^{234}U to ^{230}Th , but the measurement of multiple chronometers on the same sample improves the confidence in the final determined model age if all ages are concordant (i.e., the measured ages all agree), particularly if chronometers with different elements (and thus different chemistries) are applied. Other common radiochronometers are (progeny/parent) $^{214}\text{Bi}/^{234}\text{U}$, $^{231}\text{Pa}/^{235}\text{U}$, $^{229}\text{Th}/^{233}\text{U}$, $^{228}\text{Th}/^{232}\text{Th}$ for uranium materials and $^{234}\text{U}/^{238}\text{Pu}$, $^{235}\text{U}/^{239}\text{Pu}$, $^{236}\text{U}/^{240}\text{Pu}$ and $^{241}\text{Am}/^{241}\text{Pu}$ for plutonium bearing materials. The general method for measuring a material's age involves radiochemical separation of the small amount of the progeny isotope from the high amount of the parent isotope, followed by mass spectrometry measurements^{9,27}. The chemical separation techniques used are those familiar to many analytical chemists - ion exchange or extraction chromatography. Gamma-ray spectrometry measurements also have been applied on the original sample for non-destructive analysis of the sample's age²⁸.

For relatively pure materials, for example fuel pellets, the measured age can be regarded as fairly reliable²⁶. Radiochronometry was applied to determine the age of the seized LEU fuel pellets analyzed at ITU, discussed above, and the age of production of the UO_2 composing the pellet was determined as the end of 1990. This date fitted with the suspect manufacturer and confirmed other intelligence sources that the pellets were part of a fuel assembly stolen from the Ignalina power plant in 1992¹¹. For materials with higher impurity levels, for example UOCs, the age determination is less reliable²⁹. The model production date measured on the UOC sample seized in Australia, for instance, was considered by the authors as being anomalously old¹⁸. The sample production date was determined to be 17 January, 1964 (± 233 days). While this date was possible (it overlapped with an active period of operation of the mine/mill) the authors concluded, based on the elemental impurities (regarded by the authors as being a more reliable signature for that sample) that the sample was most likely generated in a later production period when ammonia (NH_3) was replaced by magnesia (MgO) as the precipitating reagent.

An example of a case where application of multiple chronometers did not result in concordant ages is provided by an exercise coordinated by the Nuclear Forensics International Technical Working Group (ITWG). ITWG is an organization of nuclear forensic practitioners (scientists, law enforcement and regulators) from over 30 countries. ITWG organizes and coordinates collaborative material exercises (CMX) to evaluate and improve the technical capabilities of nuclear forensics laboratories. The third exercise (CMX-3) involved two pieces of HEU metal (Figure 9). Age determination of the uranium metal samples using the $^{230}\text{Th}/^{234}\text{U}$ chronometer gave a production date of May 2003, which agreed with the date the metal was cast. Interestingly, the use of the $^{231}\text{Pa}/^{235}\text{U}$ chronometer gave a production date of 1976, i.e., 27 years older than that of the $^{230}\text{Th}/^{234}\text{U}$ chronometer. The older age measured using the $^{231}\text{Pa}/^{235}\text{U}$ system was thought to be due to the incomplete

separation of protactinium from uranium during casting. Thorium has a higher melting point than protactinium (1750°C and 1562°C respectively) and is thus more refractory; it formed insoluble inclusions that float to the top during casting³⁰. This case highlights the need for caution in the interpretation of radiochronometric results, especially when the chemistry of different progeny elements is different and may or may not have been separated from the parent to the same extent. For the CMX-3 exercise, the elemental impurities supported the radiochronometric results by suggesting the metal samples had experienced a complex processing history; the high levels of carbon (~ 1500 ppm) which is often used as mold material in the casting of HEU, as well as the presence of both zirconium and erbium, which are both used as mold coatings but not generally at the same time, was consistent with multiple castings of the uranium metal³¹.

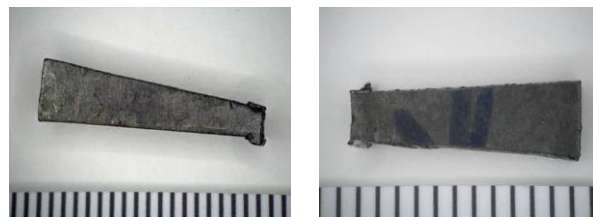


Figure 9. Two pieces of HEU metal from the third ITWG collaborative material exercise (CMX-3).

Microsignatures

Microsignatures are morphological, elemental or isotopic signatures found in either individual microscale particles or in the bulk material at a level not visible to the naked eye. Analysis of nuclear material and particles at the microscopic level adds significant value to a nuclear forensic investigation as characteristics imparted by various material processing methods may only be evident on the microscale. The microstructure of uranium, plutonium, or thorium oxide powders or UO_2 ceramic fuel pellets have been shown to be determined by both the precursor material and the hydro- and pyro- metallurgical processing conditions⁹.

The fourth ITWG collaborative material exercise (CMX-4) illustrated how microstructure can be a useful nuclear forensic signature. Participants in this exercise received two (un-irradiated) low enriched uranium (LEU) UO_2 ceramic fuel pellets and a LEU powder and were required to conduct a nuclear forensic investigation to assess the similarity between the three samples. Microstructural examination of the two pellets (e.g., grain size and porosity, see Figure 10) suggested that they were the result of different UO_2 powder feed stock, pressing or sintering conditions. Other signatures, for example uranium isotopic abundances, obtained from both bulk and microscale measurements using respectively bulk mass spectrometric techniques and nanoscale secondary ion mass spectrometry (NanoSIMS), as well as age determination, were used to provide the complete picture on the processing history of the samples³².

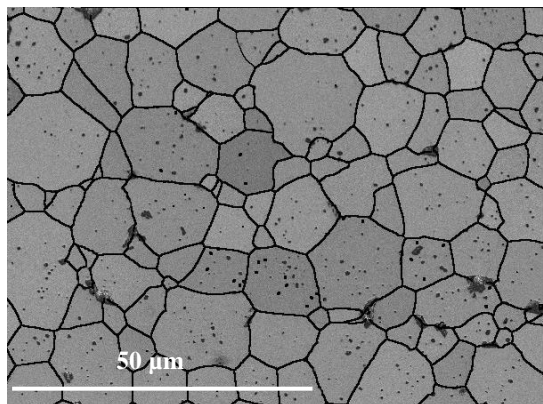
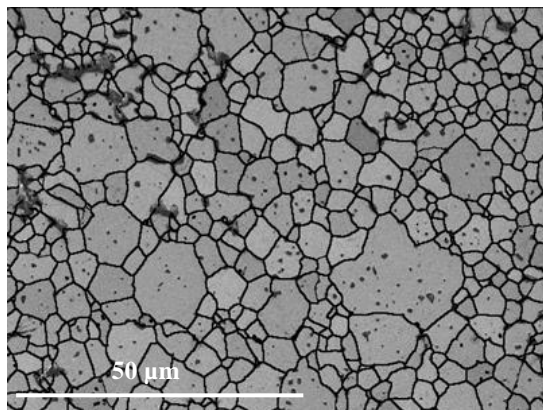


Figure 10. Scanning electron microscope maps (using backscattered electrons) of two UO_2 fuel pellets showing different grain size.

In the case of the UOC seized in Australia, discussed above, SEM examination of the powder appeared to contradict the results provided by other signatures; the microstructure (particle size, shape, surface texture) of the seized sample looked quite different to the exemplar from the suspected source (see Figure 11). The case highlights the need for detailed understanding of the microstructural changes that occur during various processing operations (such as precipitation or calcination) of different uranium products.

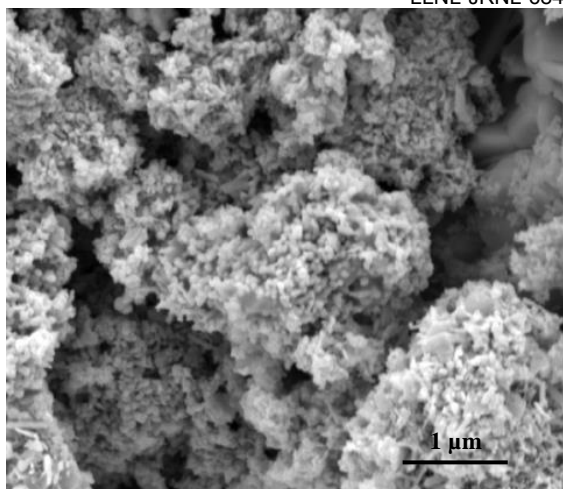
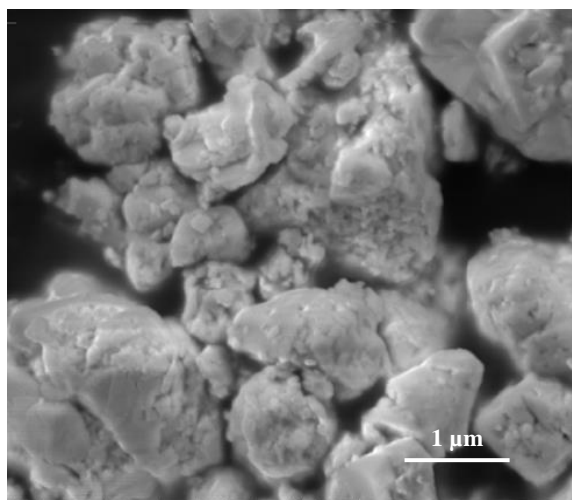


Figure 11. Secondary electron images of the seized sample from Australia (top) and a UOC powder sample from the suspected source¹⁸. Reprinted from ref 18. Copyright 2014 Elsevier.

Radioactive Materials

Whilst this paper has focused on the analysis of nuclear material, the analysis of other radioactive materials, which are widely used in industry and medicine, forms an important component of nuclear forensics. This importance is evidenced in regular media reports from around the world documenting such materials falling out of regulatory control, which are supported by data from the ITDB⁷ and also individual country's incidents registers (e.g., the Australian Radiation Incident Register (ARIR) recorded 22 incidents of missing, found or stolen radioactive sources between 2004 and 2013 in Australia³³). These materials are of particular concern due to their potential use by terrorists in radiological dispersal devices (RDD) or 'dirty bombs'. Shin and Kim³⁴ lists the key radionuclides of concern for RDD events based on criteria such as portability, relatively low security (readily obtainable), relatively high levels of radioactivity and physical and chemical form. The challenges in analyzing these materials is compounded by the high radiation doses they emit relative to those of many pre-irradiated nuclear materials, which may impose significant handling constraints.

Many of the nuclear forensic signatures applied to nuclear materials can also be utilized for other radioactive materials. In the case of commercially manufactured sealed sources, physical dimensions and identification markings, if present, are critical signatures. Frequently these characteristics, particularly when combined with a non-destructive analysis such as gamma spectrometry, may be sufficient to identify the origin of such radioactive sources even when serial numbers or other identifiers are obliterated. This is possible with the support of detailed specification sheets produced by manufacturers and comprehensive source licensing databases maintained by jurisdictional regulators.

Evaluating the irradiation and processing history of radioactive materials is an area of current research and development in nuclear forensics³⁵. Steeb et al.³⁶, for example, recently developed an analytical procedure for determining the age of radioactive strontium sources by measuring the relative amounts of ^{90}Sr and its ^{90}Zr progeny by means of chromatographic separation and mass spectrometric detection. Similarly, Charbonneau et al.³⁷, developed a procedure for determining the age of radioactive cobalt sources using ^{60}Co and its ^{60}Ni progeny. The presence of trace elements and isotopes in radioactive material may also serve as significant signatures, but will rely on the development of comprehensive databases with such information.

Traditional Forensics

As the previously given definition of nuclear forensics states, the field includes not just the analysis of nuclear or **other** radioactive material, but, particularly in cases where a relevant authority wishes to pursue a criminal investigation associated with a radiological crime scene, nuclear forensics also involves the analysis of evidence contaminated with radionuclides. The International Atomic Energy Agency (IAEA) notes that such an examination “presents a unique challenge”⁸.

For material contaminated with radionuclides, a critical consideration is whether exposure to radiation will in fact diminish the value of forensic evidence. Research has been undertaken, and continues to occur, to quantitatively explore the effects of ionizing radiation in a range of forensic evidence types including DNA³⁸⁻⁴¹, fingerprints^{41,42}, electronic devices⁴³ and fibers⁴⁴. Generally, this research suggests that these evidence types may retain forensic value in spite of exposure to ionizing radiation and thus should be examined as part of the nuclear forensics process.

The question then becomes how can such evidence be examined safely and without further compromising its evidentiary value? Forensic laboratories are generally not equipped to handle radioactive materials in any quantity. The options are either to decontaminate the evidence prior to its analysis at a forensic laboratory or to examine the item with the contamination in situ at a laboratory equipped to handle radioactive material. The decision as to which course of action to take is largely dependent on evidence type; whilst evidence such as fibers can in some circumstances be decontaminated, other types of evidence such as fingerprints are inherently unsuited to these processes⁴⁴. For DNA a ‘hybrid’ approach is adopted; research has demonstrated that extraction (the process whereby DNA is separated from the sample substrate and other cellular material, using one of a number of commercially available kits) will remove radioactive contamination, allowing the subsequent safe release of the DNA solution to a forensic laboratory for profiling^{38,45}. Forensic examinations of items with contamination in situ are generally undertaken in specialized facilities, most frequently located in nuclear institutions, where forensic and nuclear scientists can work collaboratively to obtain information from the evidence in a safe and judicially defensible manner⁴⁶.

The two cases from Australia previously described demonstrate there is enormous diversity in the kinds of traditional forensic evidence which may be encountered in the context of a nuclear forensics analysis. In the first, the UOC case, the distinctive jar in which the material was found (shown in Figure 12) was found to be a Kraft Vegemite™ jar from the period between 1956 and 1991, which was consistent with the hypothesis that the material originated from an Australian mine¹⁸. In the Victorian case, the body and leg of a moth (Figure 12) were located, and identified as being from the family *Tineidae* (fungus moths or tineid moths). Examinations are continuing in the hope that the identification of the specific species and life stage may provide information useful to determining the route by which this material fell out of regulatory control¹⁴.



Figure 12. Vegemite™ jar (left) (Reprinted from ref 18. Copyright 2014 Elsevier) and moth body (top right) and leg (bottom right) associated with seized nuclear materials in Australia.

A further case where the analysis of associated (non-nuclear) material from an incident scene was key to finding the origin of the nuclear material is that of two pieces of radioactivity contaminated metal discovered at a scrap metal yard in Karlsruhe, Germany in February 1997¹². The radioactive material was UO_2 particles (LEU and HEU) with traces of fission products; but it was analysis of the non-nuclear material that enabled the origin of this nuclear material to be determined. Elemental analysis of the metal revealed that it was stainless steel and that its origin was either Eastern Europe or Russia. The physical dimensions of the stainless steel pieces matched those of the upper and middle parts of a fuel assembly used in both an energy producing BN-600 reactor and also a BR-10 research reactor, both of which are Russian type reactors. This information, together with the nuclear material analysis, suggested that a reactor located in Obninsk, Russia, was the place of last legal control of the material.

Conclusions

Nuclear forensic science is intimately interconnected to the field of analytical chemistry; it draws analytical chemistry together with disparate disciplines ranging from sciences such as nuclear physics, geology, material science, radiochemistry and traditional forensic science, to fields such as nuclear security, nuclear safeguards, law enforcement and policy development. It is critical that the team assembled has a clear understanding of the investigative questions for which answers are sought, as this will significantly inform which nuclear forensic measurements, and the signatures that are potentially associated with those measurements, will be of value in a given case.

Though a relatively recently developed sub-discipline of forensic science, the case studies outlined in the proceeding text illustrate the important place that nuclear forensic science has come to occupy in the investigation of nuclear or other radioactive material outside of regulatory control. Unfortunately, there is no indication that the threat posed by such incidents is subsiding and thus there exists a continued need for nuclear forensic science to provide information of an appropriate standard to law enforcement and the judiciary as well as a state's nuclear security establishment.

There is currently no single nuclear forensic signature capable of identifying all unknown nuclear materials for nuclear forensic purposes. This reflects the diversity of the materials that make up the nuclear fuel cycle, from natural materials such as uranium ores to high specification processed materials such as nuclear fuels. Thus, multiple parameters need to be measured by applying multiple analytical techniques. These may include bulk analysis,

imaging and microanalysis techniques which can be used to determine the elemental and isotopic composition of a material as well as its macroscopic and microscopic structural features. It is also possible from this data to calculate properties such as the age of the material. The results, taken together, ideally characterize the sample sufficiently to narrow the range of potential origins. Corroboration of scientific measurements with sample processing history records, or sample characteristics from a nuclear forensic database, where they exist, as well as information from intelligence services, is often critical in the endeavor to answer investigative questions posed by the responsible authorities.

Effective nuclear forensic investigations rely on cooperation between international players; to deter/prevent nuclear smuggling across international borders, communication and collaboration between neighboring countries is essential. Such collaboration is facilitated through international networks such as the International Atomic Energy Agency, the Nuclear Forensics ITWG and the Global Initiative to Combat Nuclear Terrorism (GICNT). In addition, the capability to perform a comprehensive nuclear forensic analysis, if that is what is required, may not reside within a single facility or even within a single country. Pre-existing collaborations to facilitate sharing of actual samples and expertise during a nuclear forensic investigation may be crucial to the successful outcome of a real case. While the potential threat that terrorists will acquire nuclear or radiological dispersal weapons remains real, the importance of nuclear forensics as a means of providing evidence of illicit activities, particularly if seized materials are linked to more substantial quantities, is now being recognized worldwide.

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Notes

The authors declare no competing financial interests.

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ACKNOWLEDGMENTS

Lawrence Livermore National Laboratory is operated by Lawrence Livermore National Security, LLC, for the U.S. Department of Energy, National Nuclear Security Administration under Contract DE-AC52-07NA27344.

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